Progress Report and Proposed Work for Year 2

R & T 3108105-16 ONR Contract #N00014-89-J-1227

John L. Hall JILA, University of Colorado August 1995

1. **Research Description.** This ONR funding provides essential support for a vigorous ongoing research effort involving 12 people, including 3 postdoctoral researchers, 4 graduate students, and two visiting faculty members. This year, two students have earned their PhD's and left, one for industry and the other for a NRC Postdoctoral position in NIST. The focus of the work is the development of precision laser techniques and their application to physical problems of outstanding scientific interest, including laser frequency stabilization at/below the Hertz level, cold atomic "fountains", optical frequency standards, ultra-resolution spectroscopy, measurement of optical frequencies, and advanced tests of physical principles. The main thrusts in the near future are to complete our frequency measurements of the stabilized Nd laser, to exploit our new and precise spectroscopic technique based on intracavity molecular dispersion in overtone bands, to get a new atom trap for Sr atoms on line, and to bring the stable lasers together with the slow atoms.

2. **Scientific Problem**. Using the developing tools of ultrastable laser, optical frequency measurement and microKelvin atomic beam technologies we are exploring the prospects for advanced physical measurements. We believe it will soon be possible to build an optical frequency standard of unprecedented performance. We expect an optical Ramsey resonance lineshape of about 10 Hertz width at a center frequency of about 10^{15} Hertz, and the large signal to shotnoise ratio characteristic of utilizing some million atoms, namely ~1000:1. A large number of interesting scientific problems will be available with such a source. We foresee atom interferometers with frequency readout of infinitesimal Sagnac-induced or gravitational-gradient-induced phase-shifts, atomic velocity measurements at the level of a few microns/sec which lead in turn to a drastically improved limit for the charge neutrality of atoms and, potentially, a useful measurement of the gravitational response of individual atoms. Measurement of the frequency of this atomic fountain reference against the microwave frequency standard opens the door to exciting tests of our fundamental physical postulates, such as the invariance of atomic frequency ratios in time or against changes of gravitational potentials and/or fields. An exciting new possibility under active consideration is the measurement of the 4-vertex QED process which could be called "light by light scattering," but which will be measurable as a birefringence of the vacuum, induced by a string of 2 SSC magnets (6 Tesla $\rightarrow \Delta n/n = 2 \times 10^{-22}$). A collaboration with other academic and FermiLabs people has been initiated.

3. **Proposed Work: Scientific & Technical Approach.**

Our just-completed experiments with sub-microK Na atoms(see "Progress" section below) are a useful preparation for our atomic fountain atomic clock experiment. In the final stages of this planned fountain experiment, the two vertical cooling beam frequencies are changed, to provide a vertically-upward velocity reference in the molasses, thus ejecting the atomic 'pellet-like' sample upward for the atomic fountain. We are tentatively designing around the use of Strontium as the atomic system, with cooling on the 460.9 nm E_1 resonance line (reached by frequency-doubling Ti:Sapphire with a-axis $KNbO_3$), and using the ^1S-$^3P_2$ M_2 transition at 671.2 nm for the clock. This decision means that we should be free of any important "light shifts" but we must be able to do velocity selection by saturation spectroscopy on this M_2 transition. Estimates of the needed power are fairly comfortable (5 mW into a 30x build-up cavity) because of the long interaction time. As laser diodes are available now at both the needed wavelengths, it appears possible that in the future these radiations could be obtained from diode laser sources. We have gotten useful blue light levels and the glass MOT (with fritted-on sapphire windows) is just now under vacuum.

We continue to be interested in the frequency-doubled Nd laser stabilized via Iodine absorption, which offers somewhat lower performance with **vastly** lower complexity. This is probably the best of the "good enough" systems which has been demonstrated, and we have shown that a frequency reproducibility ~300 Hz *in the beat* is readily obtained. The stabilizer system built for Ti:Sapphire (external EOM and AOM, plus internal PZT for laser tuning) will be duplicated and tried as an external stabilizer for the Nd laser. The projected performance looks very good indeed, only some few milliHertz of linewidth expected relative to a reference cavity. We have gotten one set of possibly-suitable mirrors to try this locking directly at 1.06 μm. This would be useful since we could then look at the residual phase noise of the doubled green output with one of our present extremely stable reference cavities: the issue is that so far it always appears that nonlinear crystals such as used in doubling and squeezing experiments lead to excess noise. This is a good opportunity to be clear on this point which may adversely affect a number of apparently-good applications of squeezing techniques for getting low noise.

We are working toward a low-cost and attractive prospect for a practical stabilized laser for lithography and many other practical applications. Our system is an external-cavity 633 nm laser diode working with the Ne* atomic $1S_5$-$2P_8$ transition, which remarkably is only 468 GHz from the usual red HeNe standard. (We have already measured beats above 620 GHz.) We have now stabilized this diode laser to a cavity to ~10 kHz, the measurement level set by the HeNe reference used in the first experiment. A solid etalon, thermally-controlled to ~0.3 mK will offer sub-MHz stability for the stabilized diode frequency. A first-generation isolation shell for this has been constructed and is ready for testing. One may be able to have a thermally-tunable reference that can be scanned, say, 100 GHz, but yet have ~1 MHz stability.

A postdoc, Dr. J. Noh from Mandel's group in Rochester, has been working on an OPO-based scheme to generate "twin" photons for use in an interesting interferometry experiment. This idea is to follow up on a recent calculation by Burnett and Holland[1] in which amplitude-correlated beams are to be used in a Mach-Zehnder interferometer. The physical prediction is that phase can be better determined under these conditions than would be the case for coherent light. In the coherent case one expects the $\delta\Phi * \delta n = 1$ uncertainty relation to give $\delta\Phi * \sqrt{n} = 1$, whereas with amplitude-correlated light they predict $\delta\Phi * n = 1$. For a usual beam with 10^{16} quanta, this represents a fantastic change of the projected sensitivity. We now have both type I and type II OPO's working with good power conversion efficiency. The next steps are to improve our control and learn how to stabilize them, with controller noise additions comfortably at or below the shotnoise level.

4.A **Progress:**

1) Absolute Frequency measurement of Iodine-stabilized Nd laser: impediment by frequency shifts of Rb reference now overcome.
We reported this Nd absolute frequency work first last summer using an interferometric wavelength value for the reference D_2 line in Rb. Then at Christmastime we measured the frequency interval between Rb two-photon line at 778 nm and Rb D_2 d-f crossover as 1 014 234.482 ±.010 MHz. Before these last measurements there were some problems with resonance lineshape of D_2 resonance lines on hfs components with zero or only weak leakage into a third level. The theoretical and experimental situation is now better clarified: when the probe beam is intense, some assymmetry theoretically could arise as part of the lineshape modifications due to a "Berry's phase" effect, but under our symmetric sidebands condition this should not be significant. Instead, as shown in Fig 1, we have been able to account very well for the major lineshape changes in terms of momentum transfer to the atomic system by repeated absorptions in the Rb cell. A simple theory of the saturated absorption, weak probe response finds the probe beam sampling a population distribution as modified by the saturating beam, ie. atoms which are depleted in a velocity-selective way *and* whose center frequencies have been shifted by the momentum transfer from the saturating beam. The resonance lineshapes are given rather well by a series expansion of unperturbed absorbers at the unperturbed center frequency, plus ones with an increasing number of previous velocity-shifting events in their past, and correspondingly large shifts of their central frequency. Mathematically a convenient expansion parameter is $\alpha = \Omega_R \tau$, with Ω_R being the saturating pump-beam's Rabi frequency, and τ the average interaction time, basically the transit time in the low pressure domain of interest here. For the response function $R(\omega/\gamma)$ one has basically a modified Taylor's expansion formed from the Lorentzian $L(\omega/\gamma)$:

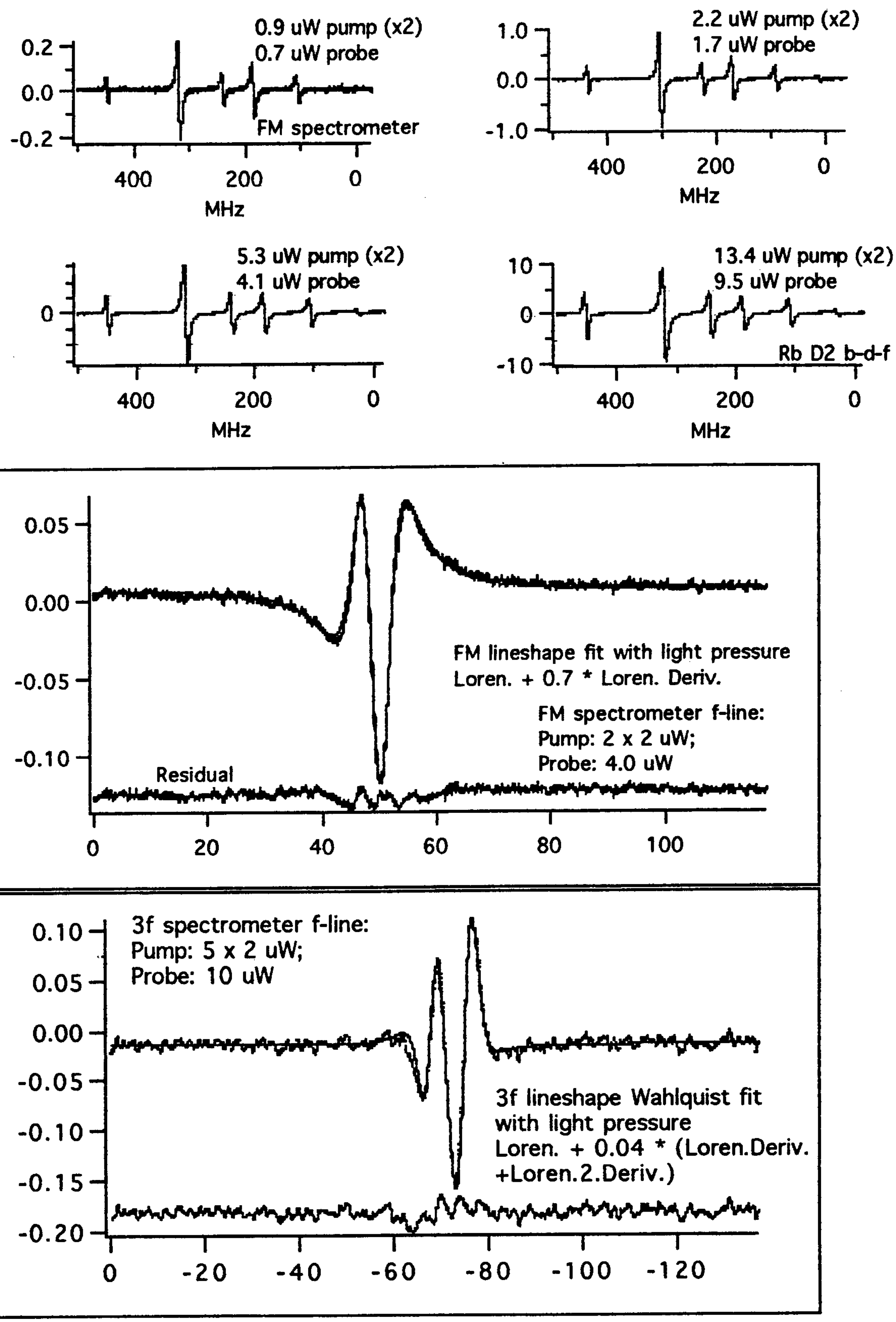


Figure 1: Rubidium saturation signals showing influence of momentum transfer.

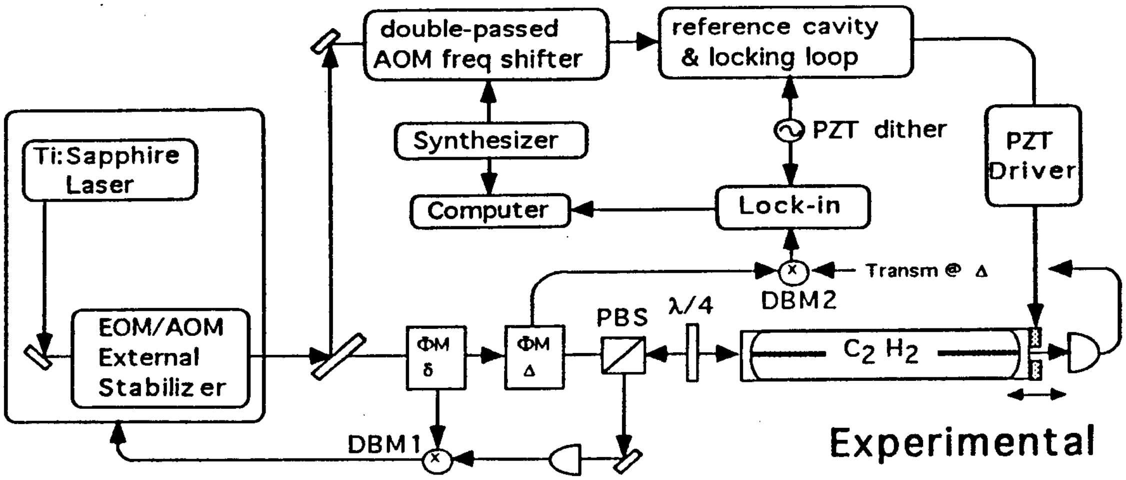
$$R(\omega/\gamma) = L(\omega/\gamma) + \sum_{n=1}^{\infty} \alpha^n \; n\Delta\omega \;\; C_n \frac{\partial^n L(\omega/\gamma)}{\partial^n \omega/\gamma},$$

where the mean recoil shift for n previous interactions is shown as n $\Delta\omega$, with $\Delta\omega$ being the recoil frequency shift per photon absorbed. The coefficients C_n are used for fitting, and should be basically unity if our model really represented the geometrical situation fully. Related expressions were given by Kazantsev, Sudutovich and Yakolev[2]. Some previous experiements by Grimm and Mlynek[3] also explored this effect in Cs, but their choice of beam sizes made it difficult to be quantitative. As may be seen from Fig 1, this simple formula is seen to be remarkably effective in clarifying the observed lineshapes!

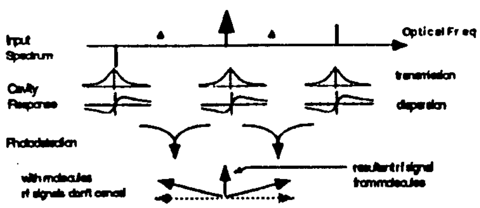
With such understanding, one can find suitable operating regimes – namely matched pump/probe powers and accurately-matched modes –such that no big shifts are expected. Evidently good high-sensitivity techniques allow use of reduced power which also is helpful. These changes made it possible to demonstrate ~2 kHz reproducibility on the Rb D2 line, comparing an rf sideband spectrometer with one based on FM "dither" and third-harmonic locking. Considering that the natural linewidth is 6 MHz, we are *accurately* splitting these tricky lines to 1 :3000. We project a "final" measurement of the Nd frequency can be performed with << 10 kHz uncertainty relative to the Rb 778 nm two-photon reference.

2) Cavity-enhanced saturated absorption molecular overtone spectroscopy offers access to thousands of reference lines of potentially sub-kHz width throughout the infrared-red-green spectrum. We have gotten narrow lines (<400 kHz transit-limited), resonances in C_2H_2 at 792 nm in the first experiments, which were presented[4] at QELS '95. Figure 2 shows some of the data now in hand. The signal has also been seen with a diode laser, but S/N was not very good in this case. Another overtone of $\Delta v=5$ is known at 643 nm and will be tried when Chris Oates' dye laser has been converted to this wavelength. An attractive - but weak - line in HCCD can be reached with the Nd 1.064 μm laser and should offer an impressive stabilization system for this laser. A special advance in this technology is noted in 4.B below.

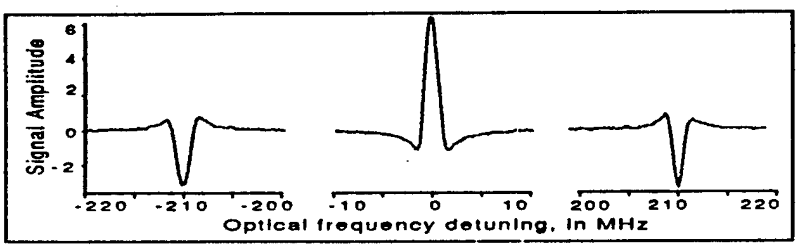
3) Chris Oates has defended his PhD thesis, the first part of which presents the uv line position measurements in Na 5P that improved the hfs parameters by 40-fold. The second part of thesis is the first precision lifetime measurement of any atomic system using *frequency-domain* techniques, and by far the most extensive study of what the *real* single atom resonance lineshape is. Measurement of the limiting linewidth at low intensity gave directly the frequency equivalent of the atomic lifetime, with an accuracy which is about ~0.2%. It is interesting that another precision experiment[5] (this one in Kaiserslautern using fast beams) has been done to address the continuuing 1% discrepancy between theory and the best previous experiment, both thought to be only <0.25% uncertain. In addition, a new theoretical result has been obrained by the group of C. Froese-Fischer. Remarkably



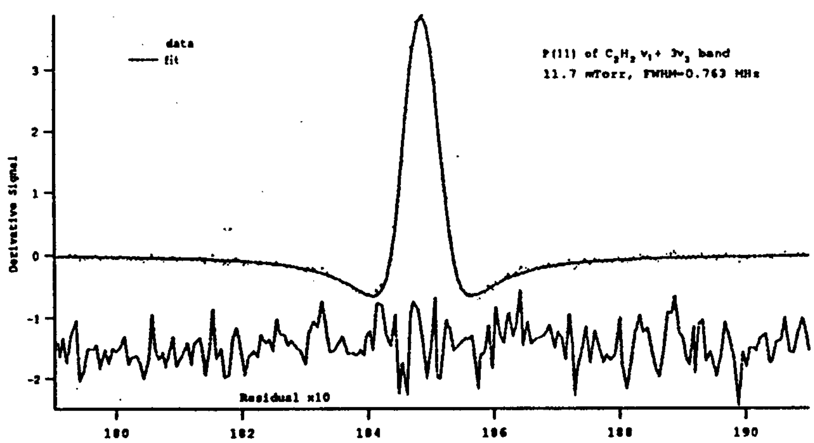
Experimental Setup



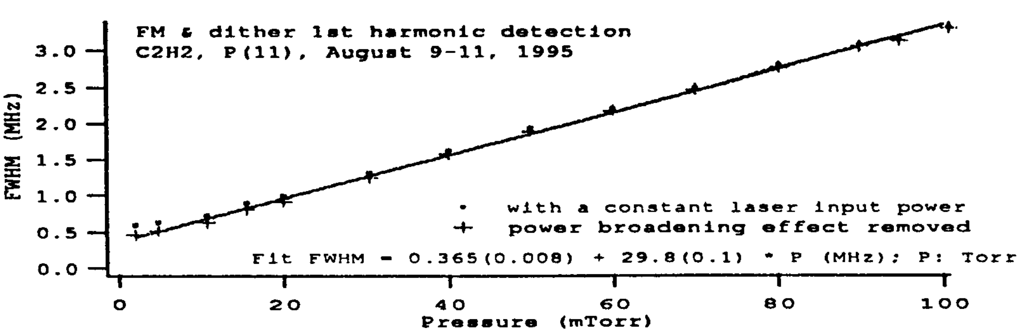
Experimental Principle



3-Peak Spectrum typical of RF Optical Heterodyne Spectroscopy



Precision Scan of Central Dispersion Peak (derivative shape)



Pressure Broadening

Fig. 2

enough, now both theoretical results agree and both modern experimental results agree, and theory and experiment are now in good agreement! Figure 3 shows the present situation. A coordinated publication in Physical Review Letters is in preparation.

4) Relative to the exciting possibility of measuring the birefringence induced by QED in a strong magnetic field, our consortium of CU and CSU researchers has submitted a proposal to NSF for infrastructure development, namely the development of an interferometer able to accurately measure birefringence in the 10^{-22} range. Unfortunately, it was not funded. An experiment proposal to FermiLabs is nearing completion to use their facilities and two SSC magnets to make the QED-induced birefringence measurements. Director Peoples of FermiLabs has expressed a great interest in having the experiment done there.

5) Cavity stabilized laser experiments by Jun Ye and Professor Ma have led to an unprecedented sensitivity level for cavity locking with low intensity red laser light: we find it is possible to measure the cavity mirrors' birefringence at the ~10 mHz level, where 1/2 wave shift would be equivalent to 541 MHz. This sensitivity is ~5 $\times 10^{-17}$. The main limitation was inadequate isolation. If we use instead a few mW level of green laser light, one finds some expected but unwelcome photorefractivity effects at the ~1 $\times 10^{-17}$ level which will require study before reaching the desired sensitivity level in the sub 10^{-22} domain. Characterizing the mirror problem vs wavelength at a few colors is an obvious idea, particularly as the problem is 5-fold larger at 532 nm compared with 633 nm.

6) In a collaboration with Ivan Getting of CIRES I have shown that it will be possible to measure absolute temperature to < 1K up to ~1000K by direct measurement of the Nyquist resistor noise. This is interesting in studying earth minerals at high temperature and pressure, as the calibrated thermocouple is pressure-deformed ~30% and likely has lost its precision calibration in the meantime. The passive resistance can be directly measured, whatever the change of its value. This thermal-calibration-free approach to temperature measurement is made possible by advances in low-noise analog amplifiers, band-limiting filters, and precision wideband multipliers. Proof of electrical performance was shown by measuring relative to a thermocouple. Experiments with digitally-processing the noise will be undertaken soon.

7) A next-generation current controller for laser diodes has been developed with 3-fold lower noise than our previous best, the circuit I developed while at CalTech. The most important additional change is the automatic crossover to a light-controlled regime when the AR-coated diode is used in an external resonator. Otherwise the diode laser power goes from μW to beyond burnout for an infinitesmal change of drive current or feedback return loss. This system

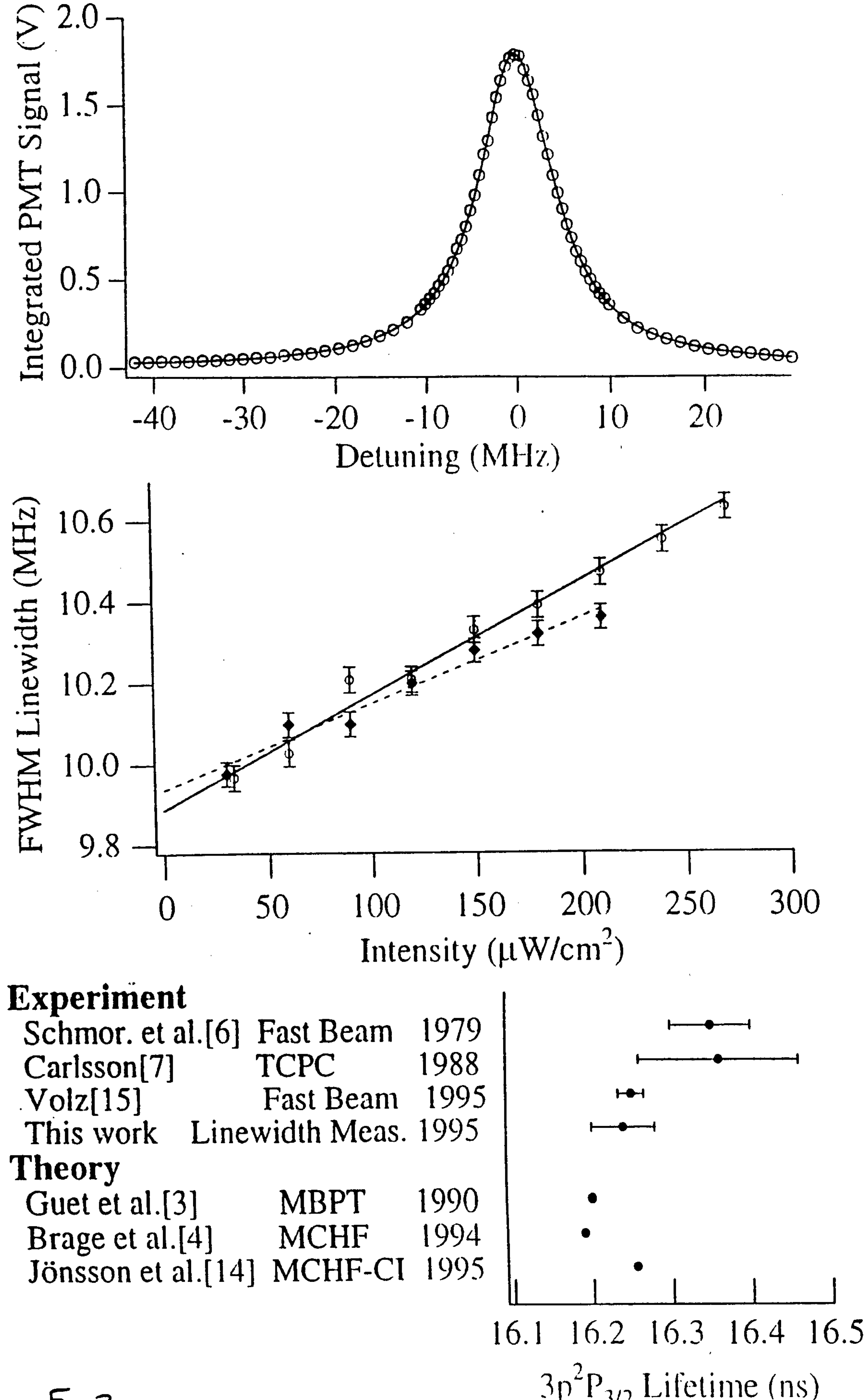

Integrated PMT Signal (V)
2.0
1.5
1.0
0.5
0.0
-40
-30
-20
-10
0
10
20
Detuning (MHz)
FWHM Linewidth (MHz)
10.6
10.4
10.2
10.0
9.8
0
50
100
150
200
250
300
Intensity (μW/cm²)
Experiment
Schmor. et al.[6] Fast Beam 1979
Carlsson[7] TCPC 1988
Volz[15] Fast Beam 1995
This work Linewidth Meas. 1995
Theory
Guet et al.[3] MBPT 1990
Brage et al.[4] MCHF 1994
Jönsson et al.[14] MCHF-CI 1995
16.1
16.2
16.3
16.4
16.5
3p²P3/2 Lifetime (ns)

Fig.3

operates near the shotnoise limit, almost ten-thousand-fold below the expensive commercial system I worked with at the Max Planck.

8) The "final" frequency locking electronic schematics for Ti:Sapphire, diode, and dye lasers are being developed for construction and general JILA dissemination. Improvements such as lower noise and dc-operated gain controls will facilitate their use by less sophisticated users. We have a reliable Ti:Sapph laser system giving 0.3 Hz laser linewidth relative to the reference cavity! Construction and installation of an appropriate reference cavity is underway now.

9) Jaewoo Noh has operated a type I OPO in $KNbO_3$ under extremely tight servo control of the cavity length relative to the pump wavelength, using a frequency-offset-locking scheme. This should make it possible to be quantitative about OPO performance in this very fragile but potentially useful system. The application will be to test the quantum noise-reduction scheme discussed relative to a Mach-Zehnder interferometer fed with correlated light beams. Burnett showed that input Foch states can lead to a $\Delta\Phi\ \Delta n$ uncertainty principle of the form $\Delta\Phi * n = 1$. The other interesting application is as a divide-by-3 module based on phase-locking the doubled 3.2 μm signal to the 1.6μm idler, thus providing a phase-coherent relationship between 1.06 μm and the 3 μm C-H stretch region where reference lines abound. We have successfully operated a type II OPO with KTP and are discussing with Professor R. L. Byer a collaboration relative to using periodically-poled Lithium Niobate crystals which can have "designer" phase-match characteristics. Unfortunately for us, there will be a "Postdoc switch-over" as Dr. Noh has accepted a nice University offer back in Korea. Dr Olivier Pfister has been the deputy for a while, so perhaps it will go smoothly enough.

4.B **Results of Special Significance**

Analysis of the sensitivity limits in intracavity spectroscopy leads to the conclusion that high finesse is desireable. However laser frequency noise is then efficiently converted into amplitude noise and the useful sensitivity is drastically compromised. We have invented an ideal detection scheme in which the full sensitivity of optical heterodyne detection is preserved with ZERO sensitivity to small laser/cavity frequency noises. The new method is called Noise-Immune, Cavity-Enhanced, Optical-Heterodyne Molecular Spectroscopy, conveniently shortened to "NICE OHMS." A patent disclosure and publication on this method are in preparation. Figure 2 top shows the configuration of this spectrometer, while Fig 2d shows a scanned line and our fit to its resonance profile. In 2e we show the measured pressure broadening of Acetylene in the $\nu_1 + 3\ \nu_3$ band near 790 nm.

5. **Extenuating Circumstances.** An exceptionally long thesis-time has been required by one of the students as he turns out to have serious problems with dyslexia. He has been very powerful in the lab and will be a good resource on electronics for an appropriate company. Unfortunately, the good PostDoc who has

been doing the OPO work has accepted an attractive (and premature) offer from a notable university back in Korea. The "deputy" postdoc, Dr Olivier Pfister, has been learning this experiment and should be able to continue the experimental program, but we will have to collaborate long distance with Dr. Noh about the theory. Another argon laser plasma tube replacement -after two last year -- is now needed.

6. **Publications**:

Papers published in Refereed Journals

1. P Jungner, S. Swartz, M. Eickhoff, J. Ye, J. L. Hall, and S. Waltman, "Absolute frequency measurement of molecular transitions near 532 nm," IEEE Transactions on Measurement and Instrumentation **44**, 151-4 (1995).

2. M. L. Eickhoff and J. L. Hall, "Developing an Optical frequency standard at 532 nm," IEEE Transactions on Measurement and Instrumentation **44**, 155-8 (1995).

3. J. L. Hall, "Frequency stabilized lasers -- from the beginning toward the future," invited submission for memorial issue honoring V. P. Chebotayev. Laser Physics (Moscow) (Jan. 1994) pp. ?

4. L. S. Ma, P Jungner, J. Ye and J. L. Hall, "Delivering the same frequency in two places: Accurate cancellation of phase noise introduced by optical fiber or other time-varying path," Optics Letters **19**, 1777 (1994).

5. H.-R. Xia, J. I. Cirac, S. Swartz, B. Kohler, D. S. Elliott, J. L. Hall, and P. Zoller, "Phase-shifts and Intensity Dependence in Frequency Modulation Spectroscopy," J. Opt. Soc. Am. B**11**, 721-730 (1994).

Papers submitted to refereed journals

1. P. Dubé, L.-S. Ma, J. Ye, P. Jungner, and J. L. Hall, "Thermally-induced self-locking by overtone absorption from acetylene gas in an external optical cavity," submitted to J. Opt. Soc. Am. B.

2. M. L. Eickhoff and J. L. Hall, "New Approaches to Real-time Precision Refractometry," submitted to Applied Optics.

3. M. P. Winters and J. L. Hall, "The Correlated Emission Laser - An Experimental Investigation," accepted by Physical Review A.

4. M. Zhu and J. L. Hall, "Short and long-term stability of optical oscillators," subm. to IEEE Trans. on Ferroelectrics, Ultrasonics, and Frequency Control.

Articles in books

1. J. L. Hall, "Frequency stabilized lasers -- a driving force for new spectroscopies," in Proceedings of the International School of Physics 'Enrico Fermi', 1992, Course CXX : *Frontiers in Laser Spectroscopy* (T. W. Hänsch and M. Inguscio, Eds., North Holland, 1994) pp. 217-239.

2. Long-Sheng Ma, Jun Ye, Pierre Dubé, and John L. Hall, "A new modulation method for sensitive nonlinear spectroscopy -- Application to molecular overtones as visible frequency references," in *Laser Spectroscopy XII*, ed. by M. Inguscio, M. Allegrini, and A. Sasso (World Scientific, Singapore, in press).

Papers published in Conference Proceedings

1. Long-Sheng Ma, Peter Jungner, Jun Ye and John L. Hall, "Accurate cancellation (to milliHertz levels) of optical phase noise due to vibration or insertion phase in fiber-transmitted light," in SPIE **2378** *Laser Frequency Stabilization and Noise Reduction* 165-75 (1995).

2. Peter Jungner, Mark L. Eickhoff, Steve D. Swarz, Jun Ye and John L. Hall, "Stability and absolute frequency of molecular iodine transitions near 532 nm," in SPIE **2378** *Laser Frequency Stabilization and Noise Reduction* 22-34 (1995).

3. C. W. Oates, K. R. Vogel and J. L. Hall, "Improved frequency-domain lifetime measurement of the Na 3p level in a magneto-optic trap," Conference on Quantum Electronics and Laser Science, QELS '95, Baltimore MD, May 21-26, 1995, Digest pp. 145-6. Invited presentation.

2. Long-Sheng Ma, Pierre Dubé, Peter Jungner, Jun Ye, and John L. Hall, "Saturation spectroscopy of molecular overtones for laser frequency standards in the visible and near-visible domain," Conference on Quantum Electronics and Laser Science, QELS '95, Baltimore MD, May 21-26, 1995, Digest page 18.

Patent Disclosures

1. L. S. Ma, P Jungner, J. Ye and J. L. Hall, "Delivering the same frequency in two places: Accurate cancellation of phase noise introduced by optical fiber or other time-varying path," submitted to NIST and CU, April 1994. NIST has not acted.

2. M. Mizushima and J. L. Hall, "An interferometer system intended to detect the anisotropy of space," submitted to NIST and CU, March 1994. Neither NIST nor the University have reacted.

7. **Unspent Funds**: We do not anticipate having unspent funds.

8. **Other Government Support**: Other funding support for this work is supplied by the National Science Foundation as a portion of the block grant to JILA for research in atomic, molecular and optical physics. The portion available for this work varies considerably and this time was sufficient to support three student salaries and some shop work; approximate total amount \$110 K. With Leo

Hollberg, we have had support for laser diode stabilization work from the AFOSR, at $85 K total per year, which has one last year to run. Ordinarily a principal source of support is the National Institute for Standards and Technology which normally supplies about $200 K equivalent for continuing research on laser stabilization and precision measurements. The remarkable recent actions by Congress leave the future of this funding in some doubt.

9. **Major Equipment Purchases**. Unfortunately one of our large-frame argon lasers is in need of a replacement plasma tube. Four modern 100 MHz scopes were obtained to replace obsolete units. A fast computer with DSP capability is needed to implement some of the active anti-vibration control strategies in software, to facilitate rapid development. As always, we are in urgent need for mirrors, Faraday isolators, AR-coated lenses, Electro-Optic Modulators etc. for the two new wavelength ranges we have started using this year.

10. **Lab Staff**: PI/Lab chief: J Hall
GradStudents: S. Swartz, K. Vogel, J. Ye, B. Tiemann
PostDocs: J. Noh, P. Dubé, and O. Pfister
Visiting faculty: Professor L. S. Ma, East China Normal University.
Professor Ennio Arimondo from Pisa joined us in November for 1 year.
Departures: M. Eickhoff and C. Oates earned their PhD's during this year.

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